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# **PCT**

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#### INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6: C09J 7/02, E06B 3/16, B44C 1/17

**A1** 

(11) International Publication Number:

WO 99/24519

(43) International Publication Date:

20 May 1999 (20.05.99)

(21) International Application Number:

PCT/US98/21613

(22) International Filing Date:

14 October 1998 (14.10.98)

(30) Priority Data:

9/307598

10 November 1997 (10.11.97) JJ

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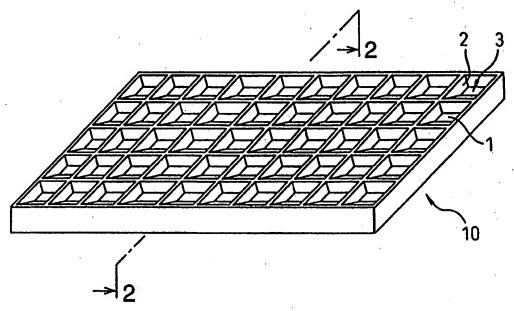
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(74) Agents: TRUSSELL, James, J. et al.; Minnesota Mining and Manufacturing Company, Office of Intellectual Property Counsel, P.O. Box 33427, Saint Paul, MN 55133-3427 (US). (81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IS, IP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, U3, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

#### Published '

With international search report.

(54) Title: ADHESIVE SHEET FOR A WINDOW GLASS STRUCTURE



(57) Abstract

A protective adhesive tape which when applied to an adherend such as window grass can simultaneously provide the effects of preventing breakage and fragment scattering, as well as heat shielding, soundproofing and vibration resistance. The tape is constructed such that protrusions made of an adhesive cross-linked polymer are arranged in a pattern on one main side of a base having two main sides, the protrusions exhibiting an elastic modulus G (at 25 °C) in the range of  $5 \times 10^5 - 4 \times 10^6$  dyn/cm<sup>2</sup> and a reduction in the log<sub>10</sub>G of less than 1.0 (at 25-125 °C), while the pattern of said protrusions can be observed through the base from the main side thereof on which no such protrusions are arranged, and the surface roughness Ra of the sections of the main side of the base on which no such protrusions are arranged is in the range of 0.4-200  $\mu$ m as measured using a contact needle surface roughness tester under conditions with a contact needle R of  $5 \mu$  and a cutoff value of 0.8 mm.

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#### ADHESIVE SHEET FOR A WINDOW GLASS STRUCTURE

#### TECHNICAL FIELD

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The present invention relates to an adhesive sheet which can be attached as a decorative sheet onto window glass and the like, for protection of privacy, prevention of glass breakage, prevention of fragment scattering and for protective effects such as heat shielding, soundproofing and vibration resistance. The present invention further relates to a window glass structure which employs this type of adhesive sheet.

#### BACKGROUND OF THE INVENTION

Most conventional adhesive sheets for window glass employ a structure with a surface base provided with decorative depressions and an adhesive layer on the back side of the base. For example, in Japanese Unexamined Utility Model Publication (Kokai) No. 6-9936 there is disclosed a protective sheet with depressions formed on a surface base for decoration, wherein the back side of the base has adhesive protrusions made from an adhesive material to prevent air leakage after attachment. However, the dimensions of such adhesive protrusions are relatively small and themselves lack decorative properties, for which reason it has been necessary to work both the base and the adhesive layer to provide the irregularities. Also, unlike the case of the present invention which will be explained in detail hereunder, this publication discloses no conditions (such as elastic modulus) wherein the adhesive protrusions prevent breakage of the window glass, or provide heat shielding or vibration resistance effects.

Japanese Unexamined Patent Publication (Kokai) No. 7-42456 discloses a heat shielding sheet for windows and other glass objects, wherein the surface base is formed from a synthetic resin sheet and a grid-like spacer is integrated with the sheet, with an adhesive applied onto the tips of the spacer. This sheet forms airtight spaces between the base, spacer and glass when attached to window glass, and can therefore provided a heat shielding effect. However, since the spacer used here consists of a relatively hard resin, it has a poor breakage-preventing and vibration resistance effect for window glass. The application step for the adhesive is also plagued with difficulties.

Adhesive sheets and the like wherein the adhesive layers themselves have irregularities are well-known. For example, in Japanese Unexamined Utility Model Publication (Kokai) No. 1-70848 there is proposed an adhesive tape comprising a support and an adhesive layer, wherein irregularities of an orderly form such as network pattern are provided on the adhesive layer for the purpose of increasing the area of adhesion onto the surface of a non-flat adherend such as wallpaper or the like. However, this publication nowhere discloses any means for providing an adhesive sheet with which breakage of the glass or other adherend or scattering of fragments of the adherend is prevented, or for providing effects of heat shielding or vibration resistance.

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Also, Japanese Unexamined Patent Publication (Kokai) No. 7-126582 discloses an adhesive tape provided with a support and an adhesive layer formed on the support having irregularities in an orderly form and arrangement, having a pitch of 1 mm or less on the surface. The irregularities on this adhesive tape have a very fine structure consisting of protrusions with a height of 20 gm or less and pits with a depth which is 10-90% of the height of the protrusions, while the adhesive comprises an n-butyl acrylate/acrylic acid copolymer and a polyisocyanate compound in a proportion of 1 g to 100 g of the copolymer. Since the irregularities of the adhesive layer of the adhesive tape are not substantially deformed even after adhesion to adherends, it is possible to distinguish letters, numerals, images and symbols through a transparent support. However, this publication discloses only means for forming such relatively small irregularities and discloses no means (such as elastic modulus) for maintaining relatively large irregularities even after attachment, in order to exhibit effects of preventing breakage of the adherend, as well as heat shielding and vibration resistance.

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Also, in International Patent Publication (WO) 95/11945 there is disclosed an article with an adhesive layer having a structural surface with microsized irregularities, which is capable of forming a space with the adherend estimated to have a volume of about 1 x  $10^3 \text{ mm}^3$ . The adhesive layer of the article is formed by ultraviolet polymerization using as the starting monomers 90 parts of isooctyl acrylate, 10 parts of acrylic acid and a relatively small amount of a crosslinking agent (0.1 part of 1,6-hexanediol diacrylate). Also, International Patent Publication (WO) 95/11655 discloses a tape having an adhesive layer with a microsized structural surface similar to the one mentioned above. Although the

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irregularities can be supported after attachment of the adhesive layer to an adherend according to the methods described in these publications, no means are disclosed for supporting relatively large irregularities after adhesion, in order to exhibit effects of preventing breakage, and of heat shielding and vibration resistance. Moreover, even though airtight spaces formed with the adherend are supported with the intent of actively utilizing them, only extremely small airtight spaces are formed.

The conventional type of adhesive sheets mentioned above can provide some degree of effect when attached to window glass, but they are not capable of achieving at the same time the effects as a protective sheet particularly desired in the technical field, namely those of improving the decorative properties, preventing breakage of glass and its scattering, and providing heat shielding, soundproofing and vibration resistance. Furthermore, they have provided no concrete suggestion of additionally providing a privacy-protecting effect (an effect of blocking the view from outside to inside when attached to window glass or the like).

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#### **SUMMARY OF THE INVENTION**

In one aspect thereof, the present invention provides an adhesive sheet comprising (i) a base with two opposite main sides and (ii) a plurality of protrusions made of an adhesive crosslinked polymer, which are arranged in a pattern on the first main side of the base, wherein when the elastic modulus G of the protrusions is measured by dynamic viscoelasticity measurement with a frequency of 1 rad/sec and in shear mode, the elastic modulus G is in the range of  $5 \times 10^5 - 4 \times 10^6$  dyn/cm<sup>2</sup> (at 25°C) and the reduction in the  $\log_{10}$ G is less than 1.0 (at 25-125°C),

the pattern formed by the protrusions can be observed through the base from the second main side thereof on which no such protrusions are arranged, and

the surface roughness Ra of the sections of the first main side of the base on which no such protrusions are arranged is in the range of 0.4-200  $\mu m$  as measured using a contact needle surface roughness tester under conditions with a contact needle R of 5  $\mu$  and a cutoff value of 0.8 mm, wherein R is a curvature radius of a tip of the contact needle used in the measurement.

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In the adhesive sheet according to the invention, the plurality of protrusions arranged on one main side of the base are preferably arranged in such a pattern that a plurality of geometrically shaped depressions are formed around them and so that the sections where no protrusions are arranged constitute the floors of the depressions. The floors of the depressions have the surface roughness Ra specified above, and are of a sufficient roughness as to cause light diffusion.

Another aspect of the present invention provides a window glass structure comprising a window glass and an adhesive sheet according to the invention attached to at least one surface of the window glass, wherein the first main side on which the protrusions of the adhesive sheet are arranged is attached to the surface of the window glass, and a plurality of discrete airtight spaces are formed by the surface of the window glass, the protrusions and the sections of the main side on which the protrusions are not arranged.

The window glass structure according to the invention may be any of a variety of the forms mentioned below, and for example, a combination of one adhesive sheet and one window glass may be used, or one adhesive sheet may be placed between two window glasses, i.e. the second main side of the adhesive sheet which has no protrusions may be also attached to the surface of a separate window glass. Two or even more adhesive sheets of the invention may also be used if necessary for the window glass structure.

# BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a perspective view of an example of an adhesive sheet according to the invention.

Fig. 2 is a cross-sectional view of the adhesive sheet of Fig. 1 along line II-II.

Fig. 3 is a perspective view of an example of a window glass structure according to the invention.

Fig. 4 is a perspective view of another example of a window glass structure according to the invention.

Fig. 5 is a graph showing the heat shielding effects of adhesive sheets according to the invention.

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#### DETAILED DESCRIPTION OF THE INVENTION

Preferred embodiments of the present invention will now be described. It is to be understood, however, that the invention is in no way limited to only the embodiments which are concretely described here.

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#### Adhesive sheet

The adhesive sheet of the present invention comprises (i) a base with two opposite main sides and (ii) a plurality of protrusions made of an adhesive crosslinked polymer, which are arranged in a pattern on the first main side of the base. The adhesive sheet of the invention is particularly suited for attachment to either or both sides of a window glass to provide a decorative function or protective effects including protection of privacy, prevention of glass breakage, prevention of fragment scattering and for protective effects such as heat shielding, soundproofing and vibration resistance.

The function of the adhesive sheet of the invention will now be explained in simple terms to facilitate understanding. First, at least one side of the base is provided with at least one continuous or non-continuous protrusion made of an adhesive polymer, and since the adhesive polymer of the protrusions is crosslinked and has a prescribed elastic modulus as mentioned above, it can be simply attached to an adherend (for example, window glass) without requiring separate adhesion means, to form relatively large spaces there between the adherend and the sheet. Furthermore, as a result of formation of these spaces, deformation or destruction of the spaces by plastic deformation (natural flow) of the protrusions and external forces is prevented, and it thus becomes possible to stably support the volume and shape of the spaces. That is, the protrusions on the main side of the base have the aforementioned prescribed elastic modulus (value of the storage elastic modulus G and degree of change in log<sub>10</sub>G), are effectively capable of elastic deformation when subjected to external force and have suitable adhesive strength, and can therefore exhibit the effects of preventing breakage of adherends such as glass, preventing scattering of fragments of broken glass, etc., as well as shielding of heat and vibration resistance.

If the elastic modulus G (value measured at 25°C) of the protrusions on the main side of the base in the adhesive sheet of the invention as measured by dynamic viscoelasticity measurement with a frequency of 1 rad/sec and in shear mode is less than 5 x

 $10^5$  dyn/cm<sup>2</sup>, the protrusions undergo plastic flow with time, making it impossible to form spaces between the sheet and the surface of the adherend. Conversely, if the elastic modulus G exceeds  $4 \times 10^6$  dyn/cm<sup>2</sup> the adhesion of the protrusions will be lower. The lower adhesion of the protrusions will tend to result in a reduction in the vibration resistance effect expected by the invention, and may also prevent achievement of airtightness at the interface between the protrusions and the adherend surface, when airtight spaces are formed between the sheet and the adherend surface.

Also, in cases where the adhesive sheet is to constitute a protective structure for window glass, the elastic modulus G of the protrusions at 25°C is preferably in the range of  $6 \times 10^5 - 2 \times 10^6$  dyn/cm<sup>2</sup>. Outside of this range it is possible that no notable improvement will be seen in the effects of preventing glass breakage or preventing scattering of glass fragments.

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Moreover, if the reduction in the  $\log_{10}G$  of the protrusions is 1.0 or greater in the range of 25-125°C, that is to say if the change in  $\log_{10}G$  in this temperature range is less than -1.0, the protrusions of the attached adhesive sheet will undergo plastic flow with time, making it impossible to support the spaces formed between the sheet and the adherend. Such plastic flow will not occur if  $\log_{10}G$  does not decrease or if it increases. However, in cases where the elastic modulus of the protrusions (i.e.,  $\log_{10}G$ ) increases, there is insufficient crosslinking of the adhesive polymer during formation of the adhesive sheet, which may cause crosslinking to progress with time, and this progressive crosslinking may result in lower adhesive strength with time. Consequently, the increase in  $\log_{10}G$  is preferably no more than +1.0.

For formation of the protrusions with the prescribed elastic characteristics described above, it is suitable to prepare, for example, a crosslinked adhesive polymer by polymerizing monomer components including an alkyl (meth)acrylate, an ethylenic unsaturated acid and a crosslinking monomer with at least 2 (meth)acrylic groups in the molecule, and doing so with the content of the crosslinking monomer in the range of 0.5-5 wt% with respect to the total monomer components. The details for such an acrylic-based adhesive polymer are discussed below.

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The surface roughness Ra of the sections of the first main tide of the base on which no protrusions are arranged (hereunder also referred to as "depressions") is in the range of 0.4-200 um, and therefore light striking those sections is scattered rendering the surface opaque or semi-transparent, to exhibit a privacy-protecting effect. If the surface roughness Ra of the floor of the depressions is too low, light will not be scattered and thus the privacy-protecting effect will be reduced, whereas if the surface roughness Ra is too large, in cases where the height of the protrusions is relatively low there will be a smaller space between the adherend and the adhesive sheet, thus reducing the other protective effects other than privacy protection. From this standpoint, then, the surface roughness Ra of the floor of the depressions is preferably in the range of 0.5-100 µm, and more preferably 0.6-50 μm. Also, the surface roughness Ra of the top faces of the protrusions, in combination with the surface roughness Ra of the depressions, is preferably to the range of 0.001-10 μm, and especially 0.005-5 μm, after attachment to the window glass, etc. This will allow the adhesive sheet of the invention to have a greater decorative effect while also exhibiting a privacy-protecting effect. In other words, when the adhesive sheet of the invention has been attached to the surface of a transparent adherend such as a window glass, the pattern formed by the protrusions and depressions is recognized as a decorative pattern by observation from the other main side of the base (side which is not adhered to the adherend) or, in the case of a light-transparent adherend, from the back side of the adherend (side on which the adhesive sheet is adhered); thus, a larger difference in the surface roughness Ra of each will result in more effective enhancement of the decorative property. Consequently, the difference in the surface roughness Ra of the protrusions and the depressions is generally preferred to be 0.5 or greater, and especially 0.75 or greater. The term "surface roughness Ra" as used throughout this specification is the value measured using a contact needle surface roughness tester under conditions with a contact needle R of 5 µ and a cutoff value of 0.8 mm.

The difference in the surface roughness Ra of the protrusions and the depressions may be achieved, for example, by subjecting only the floor of the depressions to roughening treatment by mat working or the like to render it semitransparent. Mat working involves, for example, imparting the desired surface roughness to the surface of a releasable negative frame (details given below) for preparation of the adhesive sheet in a reproducible manner.

Here, the top faces (adhesive sides) of the protrusions of the reproduced adhesive sheet may have the same roughness as the floor of the depressions. This is because they can be made so that after the protrusions have been adhered to an adherend the roughness of the top faces of the protrusions change, resulting in a different light transparency at those sections than before adhesion. Nevertheless, it is preferred for the difference between the surface roughness of the protrusions and depressions prior to adhesion to be within the ranges specified above.

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In order to form sealed, airtight spaces of the desired volume between the adhesive sheet and the surface of an adherend such as window glass when the former has been attached to the surface of the adherend, where the protrusions are arranged in a pattern around the plurality of geometrically shaped depressions, a greater effect is provided wherein vibrations and impacts applied from the second main side of the base (the side which is not adhered to the adherend) are softened before being transmitted to the adherend. In other words, the effect of preventing breakage of the glass and providing vibration resistance is improved. Such airtight spaces also serve to increase the heat shielding function.

In order to improve these different effects in a balanced manner, the volume for each airtight space is preferably determined so as to be within an optimum range. That is, the volume of each depression enclosed by the protrusions is preferably in the range of 1 600 mm<sup>3</sup>. If the volume of the depression is less than 1 mm<sup>3</sup>, the heat shielding and vibration resistance effect will tend to be reduced, whereas if it exceeds 600 mm<sup>3</sup> the effect of preventing breakage may be reduced. The volume of the depression is preferably in the range of 4-300 mm<sup>3</sup>, and more preferably 5-200 mm<sup>3</sup>.

The base of the adhesive sheet of the invention may comprise a crosslinked adhesive polymer, and thus when formed integrally with the protrusions a particularly excellent effect of glass breakage prevention and vibration resistance is exhibited. Such a sheet may also function as a double-sided adhesive sheet. In such cases, for example, a light-transparent flexible support may be attached to the second main side of the base, to readily produce a single-sided adhesive sheet. The light transmittance of the flexible support carrying the adhesive sheet is normally 60% or greater, preferably 70% or greater, and most preferably 80% or greater. The material used for the flexible support may be a

polyester resin, acrylic resin, polyolefin resin, vinyl chloride resin, fluororesin, polycarbonate resin or polyimide resin. The thickness of the flexible support is usually in the range of 10-200 µm. Also, the interface between the main side of the base and the flexible support may be subjected to simple adhesion treatment such as corona treatment or primer treatment.

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The adhesive sheet of the present invention may be an adhesive sheet formed with protrusions also provided on the second main side of the base in order to have adhesive protrusions on both of the opposite main sides, or 2 adhesive sheets may be placed together to form an adhesive sheet with a double-layered structure. In the latter case, airtight spaces may also be included in the interior of the adhesive sheet in addition to the spaces between the adherend and the adhesive sheet. This construction can be employed to easily increase the vibration resistance and breakage preventing effects.

For reference, Fig. 1 is a perspective view showing an embodiment of an adhesive sheet according to the present invention, and Fig. 2 is a cross-sectional view of the same taken along line II-II in Fig. 1.

In the case of this embodiment, as shown in Fig. 2, the base 1 and the protrusions 2 on the first main side 11 of which the adhesive sheet 10 is made are formed integrally from the same material, and spaces 3 are formed which are enclosed between adjacent protrusions. The side opposite the first main side 11 of the base 1 has been roughened at the sections constituting the floors of the spaces 3, while the opposite second main side is flat. In this embodiment, as shown in Fig. 1, the protrusions 2 on the base 1 are arranged in a grid-like pattern, thus forming rectangular depressions 3. However, as will be explained below, the pattern of the protrusions 2 and the shape of the depressions (spaces) may be any of various types for particular desired effects.

In the case of the adhesive sheet 10 illustrated here, the main side 11 with the protrusions is attached to an adherend such as window glass (not shown) by contact with its surface. Specifically, the adhesive top faces 21 of the protrusions 2 of the adhesive sheet 10 are adhered to the adherend, and the spaces 3 are thereby sealed by the adherend, resulting in formation of a plurality of discrete airtight spaces between the 3 surfaces of the sides of the protrusions 2, the first main side 11 which has been roughened and the surface of the adherend. As mentioned above, but not shown here, a light-transparent flexible

support may be attached to the second main side 12 of the base 1 to make a single-sided adhesive sheet.

#### **Protrusions**

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The protrusions formed arranged on the first main side of the base of the adhesive sheet according to the invention may form a pattern of any shape, so long as the effect of the invention is not impeded. The pattern of the protrusions is preferably one which has a roughly rectangular cross-section. A rectangular cross-section for the protrusions gives an adhesive side of sufficient area, and is satisfactory workable. Preferably, a cross-section of the protrusions gives rectangles of width 0.1-10 mm, especially 0.5-5 mm, or trapezoids of upper base 0.1-8 mm, especially 0.2-3 mm and lower base 0.2-10 mm, especially 0.5-5 mm.

#### Base

The base may be either of the following general two forms:

- (I) Consisting of the same crosslinked adhesive polymer as the protrusions and preferably formed integrally with the protrusions.
- (II) Consisting of a different material than the protrusions; specifically, a material which can be used to anchor the protrusions which are made of the crosslinked adhesive polymer, such as a plastic film, ceramic plate or metal foil, so that it can also serve as a support for the protrusions.

Although any form may be advantageously employed for carrying out the invention, as mentioned previously the preferred form is that described above in (I) wherein the protrusions and the base are integrally formed, in order to facilitate production of single-sided adhesive sheets, to increase cohesion at the interface between the protrusions and the base, and to provide a better effect of softening relatively strong impacts (i.e., an effect of vibration resistance and breakage prevention).

The thickness of the base is usually 0.05-3 mm, preferably 0.1-1 mm, and especially 0.2-0.7 mm. If the base is too thin there will be a tendency for a reduced effect of softening external forces, and when employing a form wherein the sheet is adhered to the adherend to form airtight spaces between them, there will be a possibility of lower supportability of the

airtight spaces. Conversely, if it is too thick the adhesive sheet becomes too bulky, thus reducing its ease of use and appearance when attached.

## Depressions

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The depressions created around the protrusions must form spaces (throughout the present specification these will be referred to as "airtight spaces" or "open spaces" in communication with the exterior) between the adhesive sheet and the adherend in order to exhibit the different objects mentioned above. The depressions may be arranged in any of a variety of patterns, and for example, may be arranged to form a regular geometric pattern across roughly the entire surface of the roughly flat main side of the base on which the depressions are to be formed. Otherwise, the depressions may be arranged so that the pattern formed by the protrusions and depressions is recognized as a decorative pattern (relatively complex characters or images). In either case, the height of the protrusions is preferred to be essentially uniform. This will effectively increase the adhesive strength against the adherend.

The aforementioned geometric pattern may be, for example, a lattice design, lace (diced design) or other pattern. The number of depressions provided per unit area is normally 0.3-50 per cm<sup>2</sup>, and preferably 0.5-25 per cm<sup>2</sup>. With too few depressions there is a risk of less prevention of breakage to the adherend as well as lower heat shielding and vibration resistance effects, and with too many the substantial space volume between the adherend and the adhesive sheet will inevitably be smaller than the prescribed range, which may make it impossible to achieve a vibration resistance effect.

The depth of the depressions (i.e. the height of the protrusions) is normally 0.5-10 mm, preferably 0.6-5 mm, and especially 0.7-2 mm. If this depth is too small, the vibration resistance effect may be reduced, and there may be a reduction in the vibration resistance and breakage preventing effects for the adherend.

A variety of different shapes may be employed for the depressions. Examples include geometric solids such as cylinders, prisms, pyramids, cones, truncated pyramids, truncated cones, deformed spheres, deformed ellipsoids and the like. The airtight spaces may have any one or combination of two shapes selected from the group consisting of such shapes.

Adhesive polymer

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The aforementioned protrusions are preferably formed of a crosslinked polymer with adhesive properties. The protrusions which are formed from this specified type of polymer are used to ensure adhesion of the adhesive sheet with adherends. They also provide an excellent effect of supporting the airtight spaces, when airtight spaces are formed between the adherend and adhesive sheet. In cases where the airtight spaces are deformed or lost, the air in the inner spaces:

- (a) may leak from the gaps at the adhesion interface between the protrusions and adherend surface, or
- (b) may pass through the protrusions to escape outside. The adhesive crosslinked polymer is employed for excellent adhesion, to increase the shape supportability of the protrusions and to improve air blockage (permeation resistance), i.e., for increased airtightness. Also, the elastic characteristics which are governed by the aforementioned prescribed ranges serve to provide suitable cohesive force for the protrusions and increase air blockage.

In one preferred embodiment, the adhesive crosslinked polymer is a crosslinked acrylic-based polymer prepared by polymerizing monomer components consisting substantially of an alkyl (meth)acrylate, an ethylenic unsaturated acid and a crosslinking monomer with at least 2 (meth)acrylic groups in the molecule. The crosslinked acrylic-based polymer is especially superior for airtightness. The airtightness can be particularly increased if the content of the crosslinking monomer is in the range of 0.5-5 wt% with respect to the total monomer components. If the content of the crosslinking monomer is less than 0.5 wt% there is a tendency toward lower air blockage and shape support, while if it exceeds 0.5 wt% there is a tendency toward lower adhesion. For these considerations, a particularly suitable range for the content of the crosslinking monomer is 0.7-2 wt%.

Isooctyl acrylate is preferred as the alkyl (meth)acrylate, i.e. alkyl acrylate or alkyl methacrylate, for formation of the adhesive polymer. Isooctyl acrylate imparts effective adhesion to the adhesive sides of the protrusions, facilitating attachment of the adhesive sheet. Other alkyl (meth)acrylates which may be used include any one or mixture of 2 or more alkyl acrylates or alkyl methacrylates wherein the alkyl group is one selected from

among methyl, ethyl, isopropyl, butyl, isobutyl, isobutyl, 2-methylbutyl, 2-ethylhexyl, lauryl, stearyl, cyclohexyl, isobornyl, 2-hydroxyethyl, 2-hydroxypropyl, 3-chloro-2-hydroxypropyl, hydroxyethoxyethyl, methoxyethyl, ethoxyethyl, dimethylaminoethyl, diethylaminoethyl and glycidyl.

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The ethylenic unsaturated acid is preferably (meth)acrylic acid, i.e. acrylic acid or methacrylic acid. The (meth)acrylic acid effectively increases the cohesive strength of the adhesive polymer, improves the shape support of the protrusions, and effectively increases the airtightness of the airtight spaces. other ethylenic unsaturated acids which may be used include any one or combination of 2 or more selected from among  $\beta$ -

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hydroxyethylcarboxylic acid, itaconic acid, maleic acid and fumaric acid.

The mixing ratio for the alkyl (meth)acrylate (A) and the ethylenic unsaturated acid (E) may be appropriately selected for the desired range of elasticity of the protrusions, but it is preferably in the range of A:E = 80:20-99:1.

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The crosslinking monomer with 2 or more (meth)acrylic groups in the molecule is preferably 1,6-hexanediol diacrylate. 1,6-hexanediol diacrylate effectively increases the crosslinked density of the adhesive polymer, and can provide a satisfactory balance between the adhesion, the shape supportability of the spaces and the air blockage.

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The adhesive crosslinked polymer may be obtained by polymerizing a monomer component containing the components mentioned above as the starting material, by heat or by irradiation with ultraviolet, electron or other rays. For example, as explained below, the adhesive sheet may be formed by contacting the monomer with a molding frame having the desired geometric structure and completing polymerization and crosslinking in the frame. In this case, a component which includes a monomer component containing no crosslinking monomer may be partially polymerized in advance to prepare a partially polymerized syrup with an adjusted viscosity, and a mixture of this syrup and the crosslinking monomer may then be contacted with the frame while completing polymerization and crosslinking. In this case, the viscosity of the syrup is normally 100-100,000 cps. A polymerization initiator may also be used for more efficient polymerization. The polymerization initiator may be, for example, a benzophenone-based photoinitiator (such as "Irgacure 651<sup>TM</sup>" by Ciba Geigy Inc.).

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As adhesive polymers there may be used, in addition to the aforementioned acrylicbased polymers, also rubber-based polymers such as silicone rubber, butyl rubber and styrene rubber, or polymers such as polyurethane.

Peel strength when peeled from adherends

As mentioned above, the peel strength of the adhesive sheet whose protrusions have been adhered to an adherend, when peeled from the adherend surface at 25°C, is preferably in the range of 200-2,500 g/25 mm, and especially in the range of 350-2,000 g/25 mm. If the peel strength is less than 200 g/25 mm there may be a lower effect against vibrations, while if it is greater than 2,500 g/25 mm workability may be impaired, as it may become difficult to peel off the adhesive sheet after it has been attached and positioning of the sheet will also be more difficult.

Also, when the adhesive sheet of the invention is used as a protective adhesive sheet for window glass, it is convenient if the adhesive sheet which is being used can be peeled from the glass and reattached after cleaning the window glass. From this viewpoint, it is particularly preferred for the peel strength to be less than 1,000 g/25 mm. The peel strength can be controlled by adjusting the amount of the crosslinking monomer.

#### Production of adhesive sheet

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According to a preferred embodiment of the invention, the adhesive sheet may be produced by the following reproduction method.

The monomer containing isooctyl acrylate and acrylic acid in the prescribed proportion and a polymerization initiator are placed in a stirring apparatus and subjected to ultraviolet polymerization with stirring. This step ends with partial polymerization, to a viscosity in the range of 100-100,000 cps, to prepare a partially polymerized syrup. To this syrup there are added a prescribed amount of a crosslinking monomer and additional photoinitiator, and these are uniformly mixed to prepare the starting composition for the adhesive sheet. During the polymerization, the inside of the stirrer is purged with an inert gas, usually nitrogen gas or the like.

The frame for molding of the adhesive sheet with the desired geometric structure (raised and depressed portions) is prepared in the following manner. First, a positive frame

made of relatively hard plastic is prepared to have the desired geometric structure. The plastic used may be an acrylic resin or polycarbonate resin. The release resin is contacted with the positive frame, and the ultraviolet cured release resin is released from the positive frame to give a negative frame. As a concrete example of a positive frame there may be mentioned the "Acrysandy Panel (tradename)" series by Acrysandy Co., and as a concrete example of a release resin there may be mentioned "Molding Silicon SE9555 (tradename)" by Toray-Dow Corning Co. The desired roughness is imparted to the top faces of the protrusions of the negative frame at this time. This allows the roughness to be transferred to the floors of the depressions of the adhesive sheet, so that the floors of the depressions will have the desired light transmittance. The desired roughness can be imparted to the top faces of the protrusions of the negative frame by a method wherein the floor sections of the depressions of the positive frame are provided with roughness by sandblasting or other surface polishing means. Also, after roughening the surface of the negative frame, it may be completed by coating with a fluororesin such as Teflon™. The surface roughness of the top faces of the protrusions of the negative frame is usually in the range of 0.4-200 μm.

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Next, the starting composition of the adhesive sheet is contacted with the molded negative frame and a transparent release film is used to cover the composition. Here, during the polymerization and crosslinking reaction (or, curing reaction) for the composition, the negative frame and release film are situated apart at a prescribed spacing so as to form the section for the base of the adhesive sheet and the sections for the protrusions. In this state, the release film is irradiated with ultraviolet rays from the top to complete the curing reaction, and the negative frame and release film are separated. This gives an adhesive sheet made of the cured composition.

The release film may be used for shielding of oxygen, and also has the effect of flattening the other main side of the base of the adhesive sheet. A flexible support anchored to the base, as described earlier, may also be used instead of the release film. Usually, a flexible plastic film such as polyester (PET) is used as the release film.

The following method may also be used to set the thickness of the base. That is, after casting an excess of the starting composition onto the negative frame, covering it over with the release film (or flexible support) and forming a composition-containing laminate, the laminate is passed through the gaps of a knife coater containing gaps fixed at the

prescribed spacing, and the excess portion of the starting composition is discharged from the edges of the laminate, for adjustment of the thickness and flatness of the base section.

When ultraviolet rays are used as mentioned above, the wavelength of the ultraviolet rays is usually 300-400 nm, and the irradiation intensity is in the range of 330-1,000 J/cm<sup>2</sup>.

As a different method, a glass plate of sufficient thickness may be used as the positive frame material, and the flat side of the glass plate may be engraved by the sanding method to form the desired irregularity pattern for formation of the positive frame. Also, the negative frame used may be one wherein the molding side of the metal frame has been fluororesin-treated using Teflon<sup>TM</sup> or the like.

On the other hand, the base and the protrusions may also be formed from different materials and attached together. For example, the base may be a film which has been subjected to mat working on one main side and over the entire surface rendered semi-transparent with diffusion, with the protrusions formed on the mat-worked side using the aforementioned negative frame and a monomer component which gives an adhesive polymer upon polymerization. That is, in this method the adhesive sheet is formed using the mat-worked film instead of a release film. In such cases, the base is virtually transparent at the sections where the adhesive polymer is attached (i.e., at the protrusions). It is also preferred in such cases for the protrusions to be linked (i.e. established so as to surround the depressions), for increased adhesive strength between the protrusions and the base.

## Other additives

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The adhesive sheet of the present invention may also contain various additives commonly used for pressure-sensitive adhesives, so long as the effect of the invention is not impeded. Additives which may be used here include, for example, tackifiers, plasticizing agents, ultraviolet absorbers, antioxidants, coloring materials and fillers.

#### Window glass structure

The adhesive sheet of the invention may be used to form a window glass structure for protection of window glass. The window glass according to the invention may be of any of a variety of forms, and the one produced as follows is an example thereof.

The adhesive sheet is attached in such a manner that the top faces (adhesive sides) of the protrusions contact with the surface of the window glass adherend (adherend surface), after which light pressure is applied from the non-adhesive side of the adhesive sheet by the hand or a pressure roller to adhere the adhesive side to the adherend surface. Since the protrusions contain an adhesive polymer, the adhesive sheet can be adhered to an adherend surface by light pressure application as for a common pressure-sensitive adhesive sheet. The attached adhesive sheet forms a plurality of discrete airtight spaces sealed between the depressions and the adherend surface. In such a case, the elastic modulus of the protrusions is within a prescribed range while the airtight spaces are supported from collapse by a constant air pressure, to thus increase the effect of supporting the shape and volume of the airtight spaces. The pressure application is a pressure which does not cause collapse of the airtight spaces in the process of attachment, and may be for example, in a range under 10 kPa.

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Fig. 3 is a cross-sectional view of a window glass structure produced in the manner described above. As shown here, an adhesive sheet 10 according to the invention is attached to one side of a window glass 31 with the top faces 21 of the protrusions 2 as the joining sides. Between the window glass 31 and the adhesive sheet 10 there are formed airtight spaces of relatively large volume produced by the spaces 3, to provide the desired effects described above. That is, this window glass structure can simultaneously provide effects of an improved decorative property, prevention of glass breakage and scattering of its fragments, as well as heat shielding, soundproofing and vibration resistance, while also creating an effect of privacy protection.

The double window glass structure shown in Fig. 4 can be produced as a modified form of the illustrated window glass structure. This drawing shows an adhesive sheet 10 according to the invention sandwiched between two window glasses 31 and 32. Such a double window glass structure naturally provides the same effect as the window glass structure shown in Fig. 2, but also achieves a vast improvement especially in soundproofing and vibration resistance. In addition, although not illustrated, when it is desired to effectively prevent leakage of sound from inside a room it is recommended that the window glass structure shown in Fig. 4 be positioned so that the window glass 31 is on the interior side, and the adhesive sheet of the invention attached over the window glass 31.

Incidentally, throughout the present specification the term "window glass" is used in a broad sense to include, in addition to window glass mounted for the purpose of lighting of homes and buildings, also glass mounted for lighting of various articles or erected structures, as well as for related purposes.

Examples

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The present invention will now be explained by way of examples.

# Example 1

Construction of adhesive sheet:

A partially polymerized syrup was prepared by ultraviolet polymerization from a preliminary component including a monomer component comprising 90 parts by weight of isooctyl acrylate and 10 parts by weight of acrylic acid, and using 0.1 part by weight of Irgacure 651<sup>TM</sup> (product of Ciba-Geigy Co.) as a photoinitiator. To the syrup there were added 1 part by weight of 1,6-hexanediol diacrylate as a crosslinking monomer and 0.2 part by weight of additional photoinitiator (Irgacure 651<sup>TM</sup>), and mixing to uniformity produced a starting composition for an adhesive sheet.

A glass plate with a thickness of about 5 mm was engraved by the sanding method, irregularities were formed therein in such a manner that the irregularities in the adhesive sheet prepared therefrom would be rectangular solids of the desired dimensions, and this was used as the positive molding frame for ultraviolet curing performed by the method described in the previous paragraph (Production of adhesive sheet). The irregularities were formed so as to surround depressions on the base, to obtain the adhesive sheet for this example (30 cm length x 30 cm width x 1.5 mm total height of base + protrusion). In this example, as already mentioned, instead of a release film there was used a flexible support, namely the PET film "Lumila-50TTM" (50 µm thickness, 0.01 µm surface roughness Ra) by Toray Co., which was anchored to the base after formation of the adhesive sheet.

The shape of the depressions in the resulting adhesive sheet was rectangular, the unit volume was 9 mm<sup>3</sup> (3 mm length x 3 mm width x 1 mm depth), and they were arranged at a proportion of 9 per cm<sup>2</sup> (on the horizontal plane, in a grid-like pattern). The protrusions were arranged in a grid-like manner around the depressions, and the width measured on the adhesive side was 1 mm.

# Evaluation of adhesive sheet:

The sheet of this example prepared in the manner described above was evaluated by the following method based on the parameters listed below. The results of the evaluation are summarized in Table 1.

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#### (1) Elastic modulus G

The elastic modulus G (storage elastic modulus, dyn/cm<sup>2</sup>) of the protrusions of the adhesive sheet was measured by dynamic viscoelasticity measurement with a frequency of 1 rad/sec and in shear mode. The measuring apparatus used was a "Model name: RDA II" Dynamic Analyzer produced by Rheometrix Co. The sample holder for the measurement was a horizontal plate with a diameter of 7.9 mm.

#### (2) Reduction in $log_{10}G$

The change, or reduction in the log<sub>10</sub>G was calculated from the fluctuating value against a reference elastic modulus G at 25°C read from a viscoelastic spectrum obtained in the range of 25-125°C.

#### (3) Space volume

The volume of the airtight spaces formed by the depressions was calculated from the volume of one depression of the adhesive sheet before its attachment to an object. For example, where the depression was of a shape lacking one side of a geometrical form such as a truncated pyramid or rectangle, it was calculated from those geometrical dimensions.

#### (4) Shape provision

The shape provision was evaluated based on whether airtight spaces could be formed or not. Cases where spaces (airtight spaces in this case) could not be formed between an adherend and the adhesive sheet corresponding to the depressions of the adhesive sheet when the adhesive sheet was attached to the adherend (a slide glass was used in this evaluation test) were judged as NG, and those where airtight spaces could be formed were judged as OK. The slide glass was a "MICRO SLIDE GLASS White/Green Polish No. 1 (tradename)" produced by Mitsunami Glass Industries, and had dimensions of 76 mm length x 26 mm width x 1 mm thickness.

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# (5) Shape supportability

The shape supportability was evaluated based on the change in the airtight spaces with time. After attaching the adhesive sheet to an adherend (the same type of slide glass as above was used in this evaluation test), the spaces immediately after attachment were compared with the spaces after 5 months following the attachment, and cases where virtually no change occurred were judged as OK, while those in which a clear change was seen were judged as NG. This comparison was made using 20x magnified images (taken through the slide glass) immediately after attachment and 5 months thereafter.

# (6) Breakage-preventing effect (impact resistance effect)

The breakage-preventing effect was evaluated by the falling ball impact test in the following manner. The adhesive sheet was attached so as to cover roughly the entire surface of one side of a breakable test piece (same type of slide glass as above) for use as the sample. In order to maintain a constant attachment pressure, a 2 kg-weight pressure adhesive roller was used for pressure adhesion by one pass over the adhesive sheet positioned on the test piece.

The sample prepared in this manner was positioned on an iron plate via a "FUJI PAPI (tradename)" paper towel manufactured by Taiyo Paper Industries, in such a manner that the attaching side of the adhesive sheet was facing upward, and an iron ball with a diameter of 15 mm and a mass of 30 g was dropped from prescribed heights. At the beginning of the test, the dropping height of the iron ball was 30 cm. If no breakage occurred in the breakable test piece at a dropping height of 30 cm, the height was increased to 40 cm and the falling ball test was conducted in the same manner, after which the height was increased by 10 cm each time until breakage of the test piece occurred, with the height of the previous time before breakage being taken as the result for the falling ball impact test (units = cm). The testing environment was a temperature of 25°C and a relative humidity of 60% RH.

## (7) Peel strength

The peel strength is the value of the peel resistance measured when the adhesive sheet attached to the slide glass is peeled off using a tensile tester at a pulling rate of 300 mm/min in a 180° direction under conditions of 25°C environment temperature and 60% relative humidity. However, since the adhesive sheet according to the invention is attached

to the slide glass only by the adhesive sides of the protrusions, the measured peel resistance is indicated by a maximum value and minimum value. Consequently, the measurement results given in Table 1 are shown as the maximum value (Max.), minimum value (Min.) and the average value which is 1/2 of their sum.

#### (8) Surface roughness

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The surface roughness Ra of the floors of the depressions and of the top faces of the protrusions of the adhesive sheet was measured using a surface roughness tester (SE-30K) manufactured by Kosaka Laboratory, Ltd., under conditions with a contact needle R of 5  $\mu$  and a cutoff value of 0.8 mm.

#### (9) Privacy protecting effect

The privacy protecting effect was evaluated on the basis of whether or not the state of the interior of an experimental room could be seen from outside after actually attaching an adhesive sheet of plane dimensions 30 x 30 cm to a window glass of the room. The effect in cases where the state of the interior could be seen from the outside was judged as "NO", and in that in cases where the state of the interior could barely be seen was judged as "YES".

#### Examples 2-4

The method described in Example 1 above was repeated, except that the volume of the depressions of the adhesive sheets were adjusted as follows.

Example 2: 25 mm<sup>3</sup> (5 mm length x 5 mm width x 1 mm height)

Example 3: 80 mm<sup>3</sup> (10 mm length x 10 mm width x 0.8 mm height)

Example 4: 150 mm<sup>3</sup> (10 mm length x 10 mm width x 1.5 mm height)

When adhesive sheets obtained according to these examples were evaluated in the same manner as in Example 1 above, the results obtained were as shown in Table 1 below.

Table 1

		Example 1	Example 2	Example 3	I E
Crosslinking 1 agent (pts by wt.)		1	1	1 Lixample 3	Example 4
			- 1	1	1 .
		6.7 x 10 <sup>5</sup>	6.7 x 10 <sup>5</sup>	65 105	
G (dyn/c	m <sup>2</sup> )	0.7 X 10 <sup>4</sup>	0.7 x 10 <sup>3</sup>	$6.7 \times 10^5$	$6.7 \times 10^5$
∆log <sub>10</sub> G	ì	0	0	0	0
Space vo	lume	9	25	80	
(mm <sup>3</sup> )				80	150
Shape pr		OK	OK	OK	OK
Shape su		OK	OK	OK	OK
Breakage		90	50	50	50
preventing effect				70	30
(cm)				1	
Peel	Max.	1000	880	650	650
strength	Av.	50	490	280	275
(g/25	Min.	300	100	90	80
mm)	·		·	-	
Surface					
roughnes	s Ka (μ)				
Floors of		4.0	4.2	5.2	5.7
depressio					
Top faces of		0.03	0.03	0.03	0.03
protrusion	18				
Privacy		YES	YES	YES	YES
protecting effect				1	- 20

# Comparative Example 1

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An adhesive sheet was prepared by repeating the method described in Example 1. For comparison in this example, however, a flat adhesive layer was formed to a thickness of about 1.5 mm, without adding an irregularity pattern on the surface of the adhesive sheet.

Evaluation of the resulting adhesive sheet in the manner described in Example 1 gave a surface roughness Ra of 0.03  $\mu$  and a judgment of "NO" for the privacy protecting effect for this example. Also, the value determined for the breakage preventing effect was a peel strength of 1150 g/25 mm at 30 cm, 180°.

#### Example 5

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An adhesive sheet for this example was constructed by repeating the method described in Example 1 above, except that the volume of the depressions of the adhesive sheet was adjusted to 400 mm<sup>3</sup> (20 mm length x 20 mm width x 1 mm height).

5 Evaluation of heat shielding effect

The adhesive sheets of the invention prepared for this example and Examples 1 and 3 as well as the adhesive sheet prepared in Comparative Example 1 for reference were evaluated in the following manner to determine the heat shielding effect of each sheet.

After attaching the test adhesive sheet so as to cover basically the entire side of a 50 mL volume beaker, 65°C water (30 mL) was poured into the beaker which was then allowed to stand in a room at room temperature (about 25°C). The change in temperature of the water in each beaker was measured periodically, and the temperature changes using the adhesive sheets of the invention were compared with those using the adhesive sheet with a flat adhesive surface for reference. Fig. 5 is a graph showing the results with the temperature difference (°C) from the reference adhesive sheet plotted against time (minutes).

As will readily be appreciated from the graph in Fig. 5, the airtight spaces between the sheet and the beaker with the adhesive sheet of Example 1 were able to maintain the temperature of the water to as high as about 1.6°C compared to the adhesive sheet of Comparative Example 1, after standing for 50 minutes. In the case of the adhesive sheet of Example 3, it was possible to maintain the temperature of the water to as high as about 2.2°C after standing for 50 minutes. The fact that the heat shielding effect (i.e. heat insulating effect) for Example 3 was greater than that for Example 1 is attributed to the larger space volume compared to Example 1. Also, in the case of the adhesive sheet of Example 5 it was possible to maintain the temperature of the water to as high as about 2.7°C after standing for 50 minutes, which heat-shielding (heat insulating) effect was even greater than that of Example 3. This is attributed to the fact that the adhesive sheet of Example 5 had an even larger volume for the spaces between the sheet and the beaker than in Example 3.

## Example 6

An adhesive sheet for this example was constructed by repeating the method described in Example 1 above, except that the shape and volume of the depressions of the adhesive sheet were adjusted as follows.

Depression shape: roughly square truncated pyramid (adhesive side wider)

Depression volume: 6 mm<sup>3</sup> (one side at opening of square on adhesive side = 3 mm)

The resulting adhesive sheet was evaluated in the same manner as described in Example 1, giving the results summarized below in Table 2.

# 10 Examples 7-9

The procedure described in Example 6 was repeated. However, for this example the crosslinking monomer content was changed from 1 part by weight to the amounts listed in Table 2 below. The resulting adhesive sheets were evaluated in the same manner as described in Example 1, giving the results summarized below in Table 2.

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Table 2

		Example 6	Example 7	Example 8	Example 9
Crosslinking agent wt.)	(pts by	1	0.25	0.5	2
Elastic modulus G (dyn/cm <sup>2</sup> )		6.7 x 10 <sup>5</sup>	6.0 x 10 <sup>5</sup>	6.1 x 10 <sup>5</sup>	1.1 x 10 <sup>6</sup>
Δlog <sub>10</sub> G		0	-0.4	-0.2	+0.1
space volume (mm <sup>3</sup>	B)	6	6	6	6
Shape provisions		OK	OK	OK	OK
Shape support		OK	OK	OK	OK
Breakage-preventin (cm)	g effect	80	60	70	70
Peel strength (g/25 mm)	Max.	450	1600	1000	65
	Av.	350	1000	675	50
	Min.	250	400	350	35
Surface roughness F	<b>Ra</b> (μ)				
Floors of depressions		4.2	3.3	4.1	9.3
Top faces of protrusions		0.03	0.03	0.03	0.03
Privacy protecting e	Privacy protecting effect		YES	YES	YES

#### Comparative Example 2

The procedure described in Example 6 was repeated. However, for comparison in this example, only isooctyl acrylate was used as the monomer component, and no crosslinking monomer was used. The resulting adhesive sheet was evaluated in the same manner as described in Example 1, giving the results summarized below in Table 3. With the adhesive sheet of this example it was impossible to form spaces between the adherend and the adhesive sheet.

#### Comparative Example 3

The procedure described in Example 6 was repeated. However, for comparison in this example, no crosslinking monomer was used. The resulting adhesive sheet was evaluated in the same manner as described in Example 1, giving the results summarized below in Table 3. With the adhesive sheet of this example it was impossible to maintain the spaces between the adherend and the adhesive sheet.

Table 3

	Comparative Example 2	Comparative Example 3
Crosslinking agent (pts by wt.)	0	0
Elastic modulus G (dyn/ cm <sup>2</sup> )	3.8 x 10 <sup>5</sup>	5.8 x 10 <sup>5</sup>
Δlog <sub>10</sub> G	-2	-2
Space volume (mm <sup>3</sup> )	6	6
Shape provision	NG	OK
Shape support	NG	NG

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#### Example 10

The adhesive sheet of this example was prepared by repeating the procedure described in Example 1, except that a release film (PET film silicone-treated on one side) was used instead of the flexible support.

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#### Evaluation of vibration resistance effect

The vibration resistance effect of the resulting adhesive sheet was evaluated in the following manner.

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First, 2 aluminum panels of 25 mm length x 100 mm width x 2 mm thickness were provided, and an adhesive sheet cut to roughly the same planar dimensions was sandwiched between them to adhere the 3 members together. The plurality of depressions created airtight spaces between the irregular adhesive side of the adhesive sheet and the facing side of one of the aluminum panels.

Next, a vibration detector was positioned at roughly the center of the surface of one of the two adhered aluminum panels (aluminum panels adhered to the irregular adhesive side of the adhesive sheet), on the side which was not adhered to the adhesive sheet for this example, and then the surface of the other aluminum panel (at the surface not adhered to the sheet) was plucked once with a metal rod with a diameter of about 3.5 mm, at approximately the center. The vibrations transmitted from the plucked aluminum panel to the other aluminum panel on which the vibration detector was positioned were detected with a commercially available vibration detector, an "Acceleration Pickup (tradename)" manufactured by PCB (PIEZOELECTRONICS) Co.

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Based on the detected vibrations, the damping constant hi was determined using the vibration analyzer: "3562A (product no.) Dynamic Signal Analyzer" manufactured by Hewlett Packard, Inc. Three measurements were taken, and their average was used as the measured value. The measured value for this example was 2.0, as listed in Table 4. The damping constant is the value of the first vibrational amplitude divided by the second vibrational amplitude, and thus a larger constant represents a greater damping factor, or vibration resistance effect.

# Comparative Example 4

The adhesive sheet of this example was prepared by repeating the procedure described in Comparative Example 1, except that a release film (PET film silicone-treated on one side) was used instead of the flexible support.

The vibration resistance effect of the resulting adhesive sheet was then evaluated in the same manner as for Example 10. The measured value of the damping constant h1 for this example was 1.7, as listed in Table 4 below.

# Examples 11-14

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The method described in Example 10 above was repeated, except that the volume of the depressions of the adhesive sheets were adjusted as follows.

Example 11: 1 mm<sup>3</sup> (1 mm length x 1 mm width x 1 mm height)

Example 12: 4 mm<sup>3</sup> (2 mm length x 2 mm width x 1 mm height)

Example 13: 25 mm<sup>3</sup> (5 mm length x 5 mm width x 1 mm height)

Example 14: 100 mm<sup>3</sup> (10 mm length x 10 mm width x 1 mm height)

The damping constants h1 for vibrations transmitted to the protrusions sides of the resulting adhesive sheets from their bases were measured in the same manner as for Example 10, giving the results shown below in Table 4.

Anisotropy of vibration resistance effect:

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An evaluation was then made of the anisotropy of the vibration resistance effects of the sheet obtained in this example and the sheets obtained in Example 10 and Comparative Example 4 above.

The damping constant was measured in the same manner as Example 10. In this example, however, a vibration detector was positioned on the aluminum panel adhered against the flat side of the adhesive sheet (side opposite the irregular adhesive side) to cause vibration in the other aluminum panel, and the degree of vibration transmitted from the protrusions of the sheet to the base side was measured. The damping constants (this will be referred to as "damping constant h2" for distinction from the aforementioned damping constant h1 for vibrations transmitted from the base to the protrusion side) of each example were larger than the damping constants h1. That is, the resistance against vibrations (h2) preventing transmission of vibrations applied from the protrusions to the base side was higher than the resistance against vibrations (h1) preventing transmission of vibrations applied from the base to the protrusion side, thus demonstrating anisotropy of the vibration resistance effect.

Such an adhesive sheet having an anisotropic vibration resistance effect may be utilized, for example, in the following manner. An adhesive sheet of the invention including no flexible support is used to construct a double-layered structure for a window. That is, the adhesive sheet is sandwiched between two window glass panes to adhere the three members together and construct a double window. The double window constructed in this manner is fitted in the window section of a room so that the window glass on which the protrusions are adhered faces the outside. This allows vibrations transmitted from the outside, i.e. entrance of noise into the room, to be effectively prevented. It also provides

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protection of privacy and heat shielding and prevents breakage of glass or scattering of glass fragments, while further providing a decorative property. Furthermore, if it is desired to prevent leakage of sound from the room more effectively, the window glass with the protrusions adhered thereto may be oriented toward the inside, and a separate adhesive sheets of the invention (with a flexible support) attached to the inwardly facing glass surface.

Table 4

Example No.	Space volume (mm <sup>3</sup> )	Damping constant h1	Damping constant h2
Comparative Example 4	0	1.7	1.7
Example 10	9	2.0	3.5
Example 11	1	1.9	3.0
Example 12	4	2.0	3.5
Example 13	25	2.1	3.6
,Example 14	100	2.3	4.1

It is seen from the results listed in Table 4 that the damping constant hl for Example 10 was 2.0, which was larger than the value of 1.7 for the adhesive sheet of Comparative Example 4 (having both flat adhesive sides) measured in the same manner, and therefore a greater vibration resistance effect was achieved by the presence of the spaces (airtight spaces) between the aluminum panel and the adhesive sheet. Furthermore, all of the sheets of Examples 11 to 14 also had greater vibration resistance effects than Comparative Example 4, due to the spaces (airtight spaces) formed with the adherend (aluminum panel). Effect of the Invention

As explained above, according to the present invention there is provided a protective adhesive sheet which, when attached to an adherend, particularly window glass, is capable of simultaneously exhibiting effects of improved decorative property, prevention of breakage and fragment scattering of the adherend, heat shielding, soundproofing and vibration resistance, and when attached to window glass or the like also provides an excellent effect of privacy protection. The adhesive sheet of the present invention is also noteworthy in light of the fact that it can be easily attached to adherends where needed without using separate adhesion means. The invention further provides a window glass structure which has these excellent characteristics.

#### What is Claimed is:

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1. An adhesive sheet comprising

- (i) a base with two opposite main sides and
- (ii) a plurality of protrusions made of an adhesive crosslinked polymer, which are arranged in a pattern on the first main side of said base,

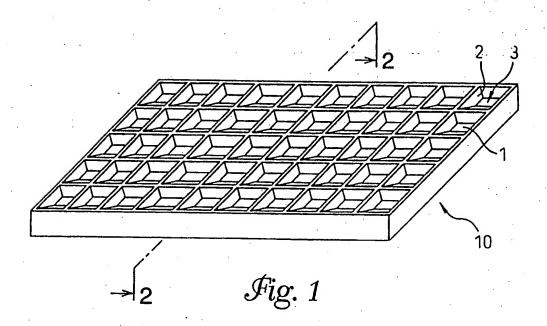
wherein when the elastic modulus G of the protrusions is measured by dynamic viscoelasticity measurement with a frequency of 1 rad/sec and in shear mode, the elastic modulus G is in the range of  $5 \times 10^5$  -  $4 \times 10^6$  dyn/cm<sup>2</sup> (at 25°C) and the reduction in the  $log_{10}G$  is less than 1.0 (at 25-125°C),

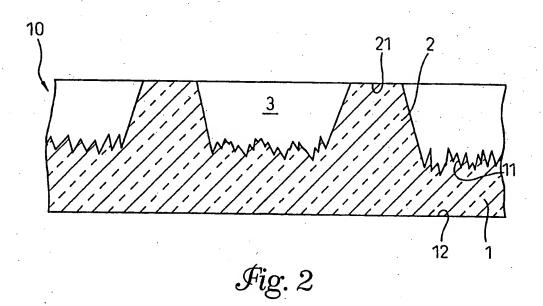
the pattern formed by said protrusions can be observed through the base from the second main side thereof on which no such protrusions are arranged, and

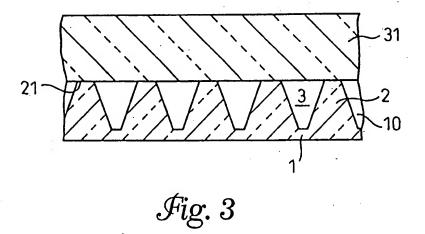
the surface roughness Ra of the sections of the main side of the base on which no such protrusions are arranged is in the range of 0.4-200  $\mu m$  as measured using a contact needle surface roughness tester under conditions with a contact needle R of 5  $\mu$  and a cutoff value of 0.8 mm.

- 2. An adhesive sheet according to claim 1, wherein the protrusions are arranged in a pattern such that they form geometrically shaped depressions around them, and the sections where no such protrusions are arranged are the floors of said depressions.
- 3. A window glass structure comprising a window glass and an adhesive sheet according to claim 2 attached to at least one surface of said window glass,

characterized in that the first main side on which said protrusions of said adhesive sheet are arranged is attached to the surface of said window glass, and a plurality of discrete airtight spaces are formed by the surface of said window glass, said protrusions and the sections of said main side on which said protrusions are not arranged.







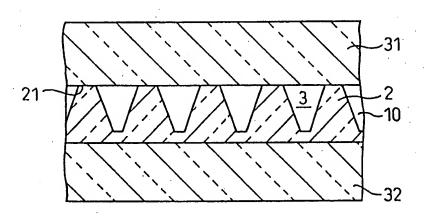
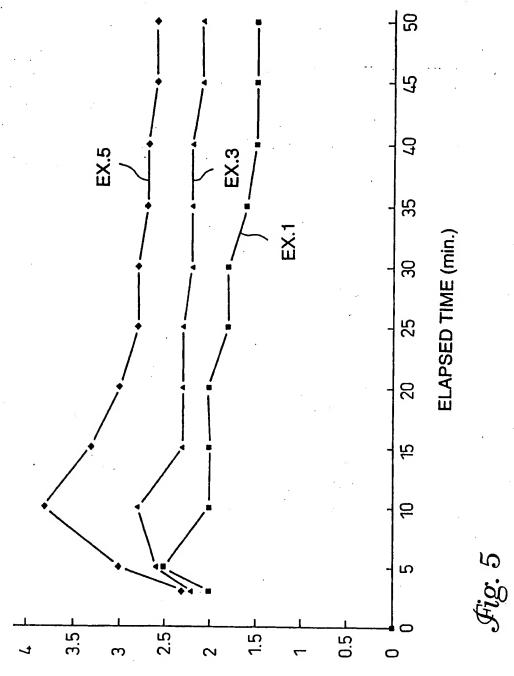


Fig. 4



DIFFERENCE IN TEMP. (°C; COMPARER TO COMP. EX.1)

# INTERNATIONAL SEARCH REPORT

Inter nel Application No
PCT/US 98/21613

A. CLASSIF IPC 6	CO9J7/02 E06B3/16 B44C1/17	,				
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	International Patent Classification (IPC) or to both national classific	ation and IPC				
	SEARCHED cumentation searched (classification system followed by classificati	on symbols)				
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Documentat	ion searched other than minimum documentation to the extent that s	such documents are included in the fields se	arched			
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C. DOCUME	ENTS CONSIDERED TO BE RELEVANT					
Category °	Citation of document, with indication, where appropriate, of the re	levant passages	Relevant to claim No.			
X	WO 97 33946 A (HATA MICHIRU ;MIN MINING & MFG (US)) 18 September		1-3			
·	see abstract					
	see page 8, line 22 - line 31 see page 9, line 29 - page 10, l	ine 7				
	see page 17, line 1 - line 4		2 - 1			
ļ	see page 17, line 9 - line 10					
Ì	see page 20, line 3 - line 9		•			
	see example 1 see claims 1,2,5					
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Furt	her documents are listed in the continuation of box C.	χ Patent family members are listed	in annex.			
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# INTERNATIONAL SEARCH REPORT

...formation on patent family members

Inter: al Application No
PCT/US 98/21613

		101/03	30/21013
Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9733946 A	18-09-1997	JP 9241589 A AU 2205397 A	16-09-1997 01-10-1997